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ORIGINAL ARTICLE

Thermal conditions for the formation of self-assembled cluster of droplets over the water surface and diversity of levitating droplet clusters

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Abstract
The effect of temperature profile of the surface of a water layer on the formation and geometrical structure of a cluster of levitating droplets is studied in a series of laboratory experiments. The experiments show that a local temperature maximum of the water surface is a necessary condition for the droplet cluster formation. A quantitative criterion for the transformation of a monolayer of randomly placed microdroplets into a self-assembled cluster of relatively large droplets is obtained. A qualitative physical description of the formation of a flat levitating droplet cluster of an axisymmetric hexagonal structure is given, based on the experimentally verified concept of the decisive role of aerodynamic forces acting on water droplets from an upward vapor-air flow. Unusual droplet clusters resulting from high surfactant concentrations and rapidly changing or more intense local heating of the underlying water layer were observed. These are elegant ring clusters, small clusters of a controlled number of large droplets, and chain clusters with branching chains of droplets in their central part. The use of a recently developed experimental procedure based on the injection of initial microdroplets with a piezoelectric dispenser makes it possible to generate hierarchical clusters, which contain continuously transforming aggregates of several droplets held in contact by electrostatic interaction. An overview of various types of droplet clusters including relatively stable hexagonal clusters, emerging and rapidly breaking up ring-shaped clusters, small clusters of a desirable number of nearly identical droplets, chain clusters containing growing branched chains of water droplets, and hierarchical clusters with rearranged small groups of nearly merging droplets provides a complete picture not only of the transition from chaotically moving droplets to self-arranged clusters, but also of the diversity of droplet clusters.

Keywords Droplet cluster · Levitating droplets · Transforming clusters · Experiments · Physical modeling

Highlights
- New experiments on the formation of clusters of levitating droplets are presented..
- A quantitative criterion for the formation of self-arranged clusters is obtained.
- A physical description of the evolution and transformation of clusters is given.
- A variety of droplet clusters observed in experiments are described.
- The use of levitating droplets as microreactors in biochemical studies is considered.

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1 Introduction

A layer of microdroplets over a uniformly heated surface of water [1, 2] and a droplet cluster levitating in an air-vapor flow over a locally heated area of the surface [3] differ significantly in their dynamics and structure. In the first case, the droplets are positioned randomly and the effects typical of a droplet cluster [4] are not observed: the self-assembly of an ordered structure, condensational growth of droplets, as well as displacement of the cluster as a whole while maintaining its structure. The relatively strong heating of the water surface is the main factor responsible for the formation of droplet clusters. The fundamental importance of local heating for cluster formation is well known [5–7], but the transition from chaotically moving droplets to a cluster remains poorly understood. However, it is clear that with fixed maximum

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water temperature and ambient air properties, the condition for the formation of a droplet cluster is a small area of a hot spot on the water surface [6, 7]. The ordering of the droplets can be described by the so-called Voronoi entropy [8, 9]. As a result, the intuitive difference between the regular arrangement of the droplets and the chaotic one was quantified.

The main objective of this paper is to provide quantitative experimental results on the effect of relatively strong local heating of the water surface on the possible formation of self-organized droplet clusters and to determine the critical condition under which a droplet cluster is formed. The understanding by the authors of the physical mechanism for the self-assembly of a drop cluster and the formation of a hexagonal structure with the largest drops in the center of the cluster is presented.

In addition to a specific physically meaningful result, we would like to give interested colleagues more information about the different droplet clusters that the authors were able to obtain in a series of experiments with varying parameters of the problem. Therefore, also discussed are clusters, which are unusual in shape and structure, obtained in laboratory experiments over the past few years. The collection of new images, accompanied by minimal explanation, complements the detailed analysis published in recent journal papers and provides a complete picture of the diversity of the observed droplet clusters.

It should be noted that so-called small clusters of almost identical droplets considered in Sect. 4.2 seem to be suitable for the possible use of levitating droplets as microreactors for studying chemical and biochemical processes. At the same time, the merging of neighboring droplets characteristic of hierarchical clusters (Sect. 4.4) is expected to be used to observe the reactions of different chemicals in these droplets. This is a preliminary view of the practical application of droplet clusters.

2 Laboratory setup and experimental procedure

The experiments were performed in the laboratory of microhydrodynamic technology at the University of Tyumen (Russia). The schematic of the experiment [10, 11] is presented in Fig. 1a. A droplet cluster (1) is formed over a locally heated area of a layer of distilled water (2) containing the surfactant impurity (sodium dodecylsulfate at a concentration of 0.5 g/L [9]). A sitall substrate 400 μm thick (3) is glued to the metal bottom of the cuvette. The cuvette body has channels (4) connected to the external circuit of the CC 805 cryothermostat (Huber, Germany), which allows stabilizing the temperature of water layer in the range of T_0 from 9 °C to 60 °C. The water layer is locally heated by a

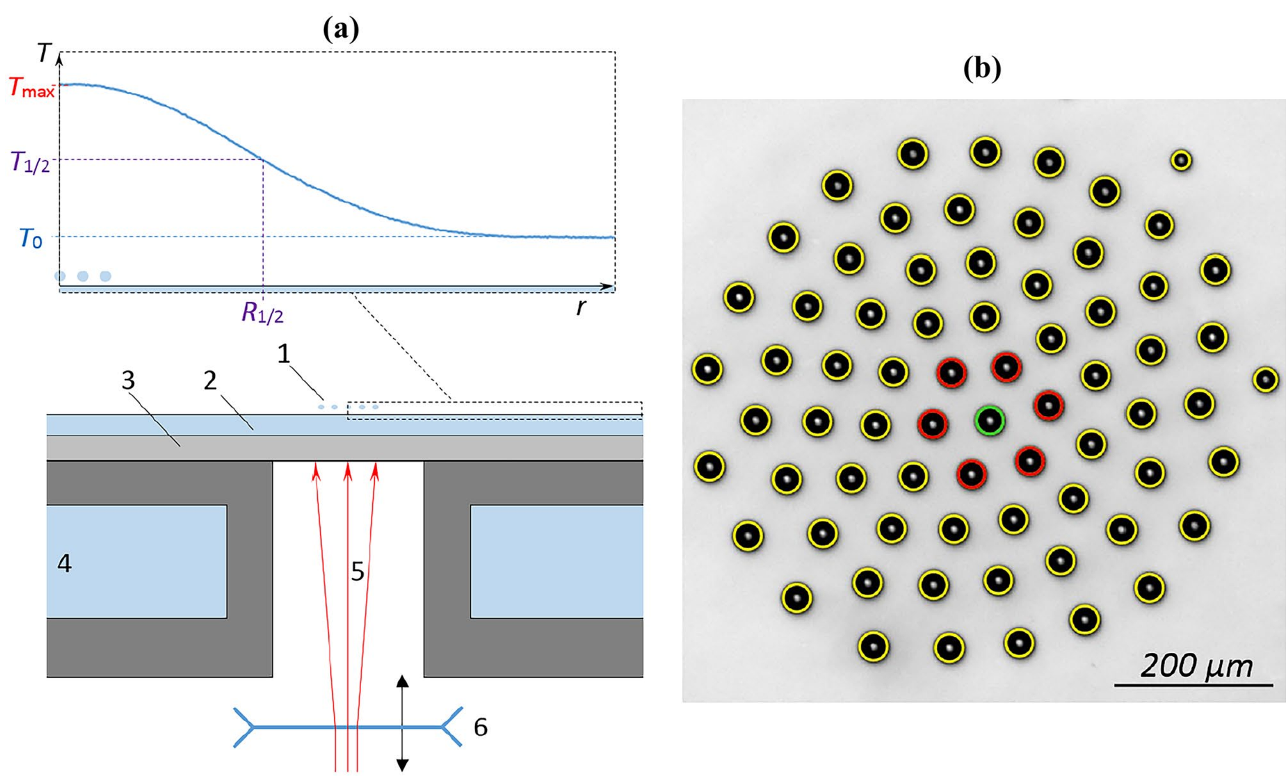


Fig. 1 **a** The schematic of local laser heating of water and **b** the top view of droplet cluster

laser beam (5) directed to the lower blackened surface of the substrate. The semiconductor laser BrixX® 808-800HP manufactured by Omicron Laserage (Germany) is used. A displacement of the defocusing (concave) lens (6) with a focal length of 10 mm made it possible to vary the diameter of the hot spot. In all experiments, the thickness of water layer was equal to 400 ± 2 µm. The axisymmetric temperature field of the water surface was recorded with an A655sc thermal imager (FLIR, USA). Video recording of the cluster was carried out using an AXIO Zoom.V16 stereomicroscope (Zeiss, Germany) equipped with a PCO.EDGE 5.5C high-speed camera (PCO, Germany).

3 Experimental results and discussion

The experiments under discussion were performed with a steady-state surface temperature profile of the water layer but without the external infrared irradiation used in [12, 13] to stabilize the droplet cluster. All temperature profiles considered were axisymmetric, with a maximum in the center and a monotonic decrease in temperature with distance from the axis. The parameter $K = T_{1/2}/R_{1/2}$ (where $T_{1/2} = (T_{max} - T_0)/2$, $R_{1/2}$ is shown in Fig. 1a) was used as the characteristic of the temperature profile in the analysis of experimental results. Figure 1b shows a typical image of a cluster with automatically recognized droplets. The experimental study included two stages described below.

3.1 Transition from a monolayer of droplets to a droplet cluster

In a series of experiments, the above introduced parameter K decreased due to an increase in the thermostating temperature T_0 at a constant maximum temperature T_{max} = 60 °C. In each experiment, the time required for cluster formation was maintained. Figure 2 shows two typical images of clusters and the corresponding Voronoi tessellation illustrating the ordering of droplets in the cluster. The Voronoi entropy is defined as $S = \sum_{n=1}^{N} P_n \ln P_n$, where n is the coordination number of a polygon, which is the number of sides of a Voronoi polygon, and P_n is the fraction of the number of polygons having the coordination number n. To generate the Voronoi tessellation, we used the free MATLAB code

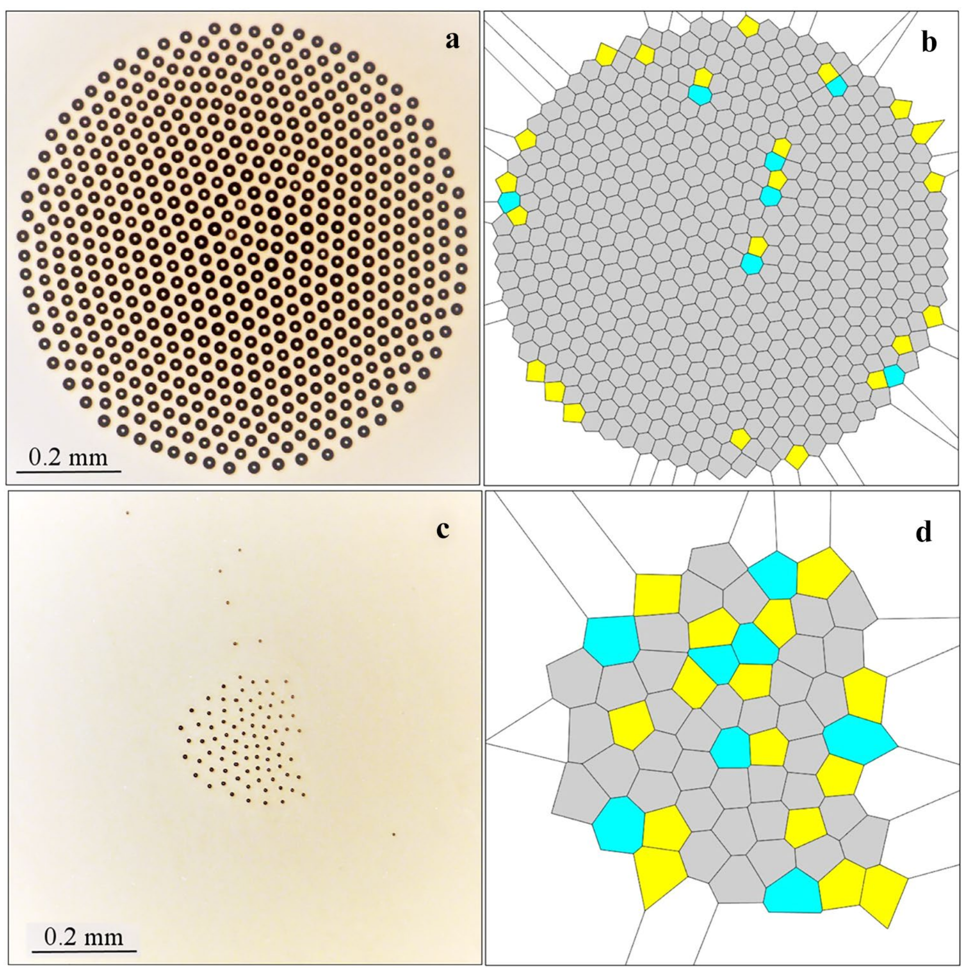


Fig. 2 Change of droplet cluster with the parameter K: **a**, **b** – K = 17.2 K/mm, S = 0.24, **c**, **d** – 2.9 K/mm, S = 0.91; **a**, **c** – images of clusters, **b**, **d** – Voronoi tessellations (pentagons, hexagons, and heptagons are highlighted in yellow, gray, and blue, respectively)

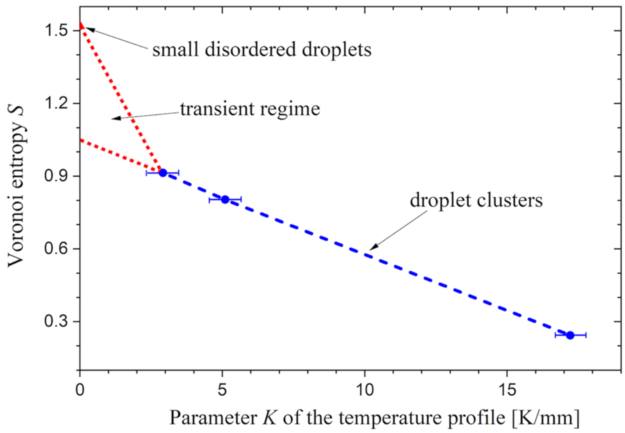


Fig. 3 Effect of local heating of water surface on ordering the levitating droplets

developed by the Department of Physics and Astronomy at the University of California, Irvine, which is available at https://www.physics.uci.edu/~foams/do_all.html, updated for our needs.

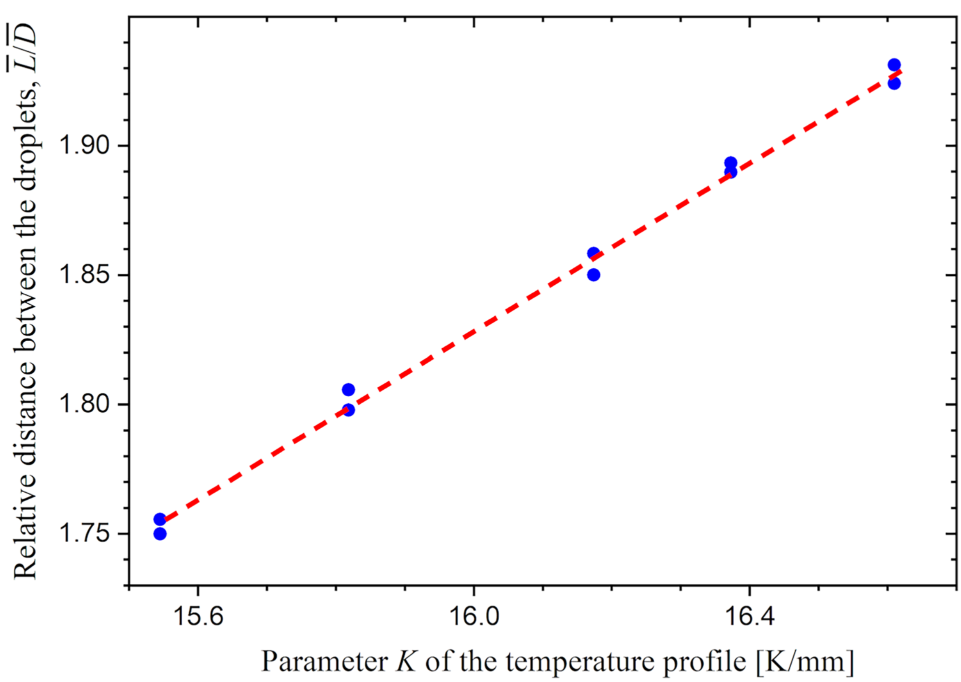
The transition from a monolayer of droplets to a cluster was observed in the range of $1\frac{\mathrm{K}}{\mathrm{mm}} < K < 2.5\frac{\mathrm{K}}{\mathrm{mm}}$. This regime shown in Fig. 3 is characterized by a significant decrease in the Voronoi entropy to values typical of regular clusters of water droplets. At larger values of the parameter K, the Voronoi entropy decreased linearly with the increase in K. Note that the orderliness of the cluster can also be estimated using the so-called continuous measure of symmetry. The definition of the continuous measure of symmetry and the details of its correlation with the Voronoi entropy can be found in paper [14].

3.2 Effect of the hot spot area on the density of droplets in a cluster

The experimental set-up enabled us to change parameter K at constant temperatures T_0 and $T_{\max}$. For this, vertical displacements of the lens were used (see Fig. 1a), and a small change in $T_{\max}$ was compensated by adjusting the laser heating power. In the series of experiments at $T_0 = 29 \pm 0.2$ °C and $T_{\max} = 70 \pm 0.2$ °C, five different lens positions were used, corresponding to the range of $15.5\frac{\mathrm{K}}{\mathrm{mm}} \le K \le 16.6\frac{\mathrm{K}}{\mathrm{mm}}$, when water droplets form a classical hexagonal cluster. The images of the cluster were processed by a special computer code that allows measuring the position and diameters of the droplets, as well as calculating the average values of the droplet diameter $\overline{D}$ and the distance $\overline{L}$ between the centers of neighboring droplets in the selected group. In Fig. 1b, yellow circles indicate automatically tracked droplets. The central droplets of the cluster, circled in red, were used when measuring the average distance between the droplets. The averaging was performed over this group of seven droplets. The ratio of $\overline{L}/\overline{D}$ was used to evaluate the density of droplets in a cluster.

The results for clusters with the number of droplets from 60 to 80 and the average diameter of central droplets from 31 to 33 μm from two video recordings for each of the five clusters are shown in Fig. 4. In other words, ten video recordings were analyzed. The temperature measurement error declared by the manufacturer of the thermal imager is

Fig. 4 Effect of parameter K on the ratio $\overline{L}/\overline{D}$, which characterises the density of droplets in a cluster. The strait dashed line shows that the dependence of $\overline{L}/\overline{D}$ on K is almost linear



2%, and the accuracy of measuring linear dimensions on the video recording frames was limited by the size of the image pixel, which resulted in an error of ± 0.4 μm.

It is seen in this figure that the distance between the neighboring droplets in a cluster is sensitive to changes in K even in the case of a constant temperature of the water surface under the cluster. As one might expect, a decrease in the area of the hot spot leads to an increase in the gas flow rate and to a decrease in the density of droplets in the cluster.

Analysis of conditions for formation of a levitating droplet cluster would be incomplete without a physical model of cluster evolution based on numerous laboratory observations, which are cited in the appropriate parts of the paper. At significant local heating of water surface, when conditions for droplet cluster formation arise, water evaporates intensively and an upward gas flow from a mixture of water vapor and air is formed. In other words, we are dealing with a flow of humid air, not just water vapor. This statement becomes clear when one recalls that the pressure of saturated vapor, even at 80 °C, is less than 50% of the normal atmospheric pressure. Since the room temperature is much lower than the humid air temperature at the water surface and the air contains dust particles, the water vapor, when cooled, condenses on these particles and small water droplets are formed. Some of these droplets are carried away by the gas flow (then, when mixed with the main flow of relatively dry ambient air, these droplets evaporate), other droplets manage to become larger due to condensation of supersaturated vapor and descend by gravity, approaching the water surface. The droplets cannot escape the flow of humid air, because the pressure of the surrounding still air is noticeably higher. Moreover, the falling water droplets collect in the central zone of the flow, where gas velocity is higher and pressure is lower, forming the basis of a future cluster.

Water droplets cannot remain at a significant height, because then they will be carried away by the gas flow and evaporate. As a result, a relatively thin cloud of a large number of droplets is formed. The larger droplets in the central part of the droplet cloud are responsible for the local increase in the velocity of the gas flow around them, so droplets located at a greater distance from the axis tend not only to remain in the same plane as the larger droplets, but also to be as close to them as possible. Thus, the cloud of droplets gathers into a flat cluster. The distance between the droplets is determined by the gas flow rate (see details in Sect. 4.3).

It is known that a droplet cluster continues to grow both due to joining of peripheral small droplets and due to vapor condensation, which leads to an increase in the size of the central droplets. There comes a moment when the most massive droplets coalesce with the water layer and disappear. Typically, the diameter of the droplet increases at a rate of about 1 μm/s, which limits the cluster lifetime to a few tens of seconds. However, for example, external infrared irradiation can completely suppress condensation growth and then the cluster lifetime is limited by the period of maintaining the right conditions [12, 13, 15].

The above description of cluster evolution is based on the concepts of gas dynamics. At the same time, it is known that evaporating water droplets have their own electric charge, and therefore, the behavior of a droplet cluster can manifest the electrostatic interaction of droplets. In this regard, we should refer to papers [16–19], in which a purely electrostatic model of the structure of a droplet cluster is considered. At the same time, it was shown in [20, 21] that the parameters of the cluster are determined by aerodynamic forces and the presence of a small electric charge of water droplets (several hundred of elementary charges) is a minor factor. However, electrical effects should be taken into account when analyzing the behavior of droplet aggregates in hierarchical clusters discussed below. Interestingly, in a recent experimental study [21], the authors of the present study were able to approximately determine the charge of the droplets, which is very close to that previously obtained by the proponents of the electrostatic model.

4 Formation of diverse clusters of levitating water droplets

It is interesting that the change in the properties of a thin surface layer of water, the variation of intensity of local heating of water, as well as the use of special devices for independently generating droplets placed above the heated area of the water surface can be used to obtain a wide variety of droplet clusters of different size, shape, structure, and behavior over time. For a general idea of the possible options, some amazing examples of such clusters are demonstrated below. The experimental details and theoretical explanations of the observations can be found in recently published papers [22–26].

4.1 Formation of a ring-shaped cluster

It turns out that the droplet cluster is very sensitive to thermocapillary flows in the thin surface layer of water below the cluster. These flows may form due to significant temperature gradients in the locally heated spot on the layer surface [27, 28]. A typical, relatively stable, droplet cluster is only obtained with natural or specially added surfactants that suppress thermo-capillary flows. In [22] it was shown for the first time that in the absence of surfactant the ordinary cluster collapses, and at a certain stage of this process an unusual ring-shaped cluster is formed. One of the images of this amazing cluster during the beginning of the formation of its second ring is shown in Fig. 5.

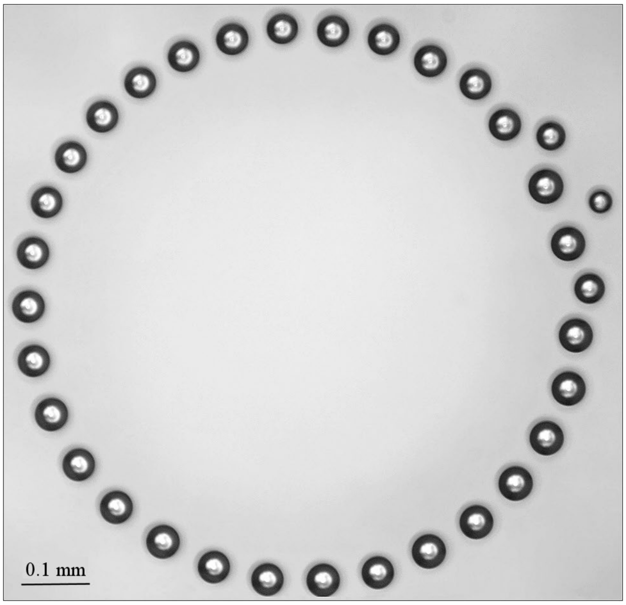


Fig. 5 A photograph of a ring-shaped cluster being formed

A ring of droplets surrounds the toroidal vortex area. As a rule, only one ring is formed. However, it depends on the number of droplets in the cluster, and it is possible to form both an open ring (an arc if there are few droplets) and more than one ring if the number of droplets is large.

4.2 Small droplet clusters

Droplet clusters usually contain a large number of droplets of various sizes. At the same time, considering the possible use of levitating droplets as microreactors for studying chemical and biochemical processes, it would be better to have clusters of a small number of the largest droplets of almost the same size. It turns out that such clusters, called "small clusters", can be generated using a simple method developed and implemented by the first author of the present paper. For this, at first, a relatively low power of local heating of water is used, which makes it possible to obtain a droplet cluster from a large number of small droplets. These droplets migrate continuously to the center of the cluster. Upon reaching the desired number of droplets in the central zone, the heating power increases sharply (for example, 2–3 times). As a result, a much more intense ascending steam–air flow carries away the smallest droplets from the cluster periphery, while the central droplets increase in size and become nearly identical. A description of the experimental method and the equipment used can be found in paper [23].

Images of some small clusters (in addition to those published) are shown in Fig. 6. It should be noted that small clusters have a structure that differs from the classical hexagonal structure of ordinary large clusters. Classification of small clusters according to their symmetry was reported in [25]. It is interesting that using the method described above, it is possible to generate small clusters with a specified number of droplets from one to several dozen. Moreover, starting from 33 droplets, the variety of cluster structures degenerates into a universal hexagonal structure, which does not depend on the number of droplets [25]. It is important to note that small clusters are very convenient objects of special studies.

4.3 A transition from hexagonal to chain cluster

Droplet clusters like the one shown in Fig. 2a are best known. Because of the mutual arrangement of the nearest droplets, such clusters are called hexagonal. It turns out that the structure of the central part of an ordinary hexagonal cluster consisting of a large number of droplets changes with a significant increase in the local heating of the water layer [24]. This change is explained by an increase in the flow rate of the mixture of water vapor and air under the center of the cluster. Recall that the height of cluster levitation above the water layer is approximately equal to the diameter of large droplets, that is, almost two orders of magnitude less than the diameter of a large cluster. Therefore, the water vapor formed under the central part of the cluster cannot go around

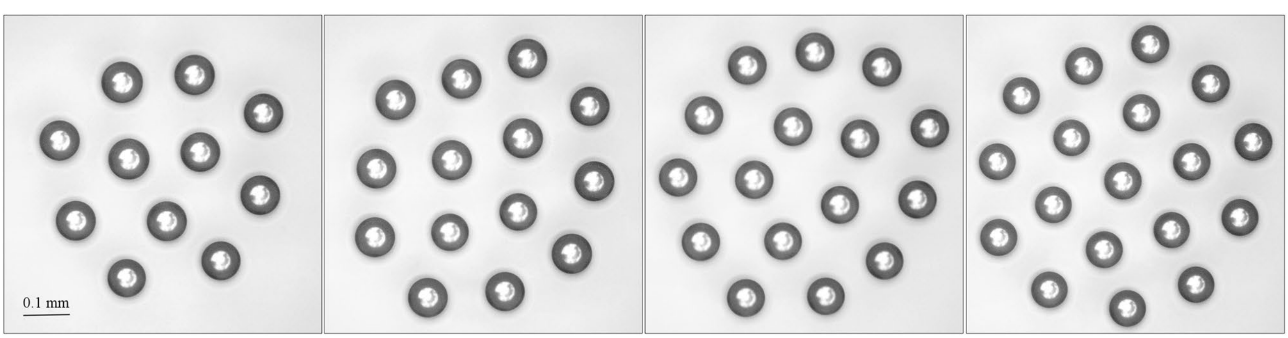


Fig. 6 Typical small clusters of nearly identical droplets. Clusters of 11, 14, 16, and 18 droplets (from left to right)

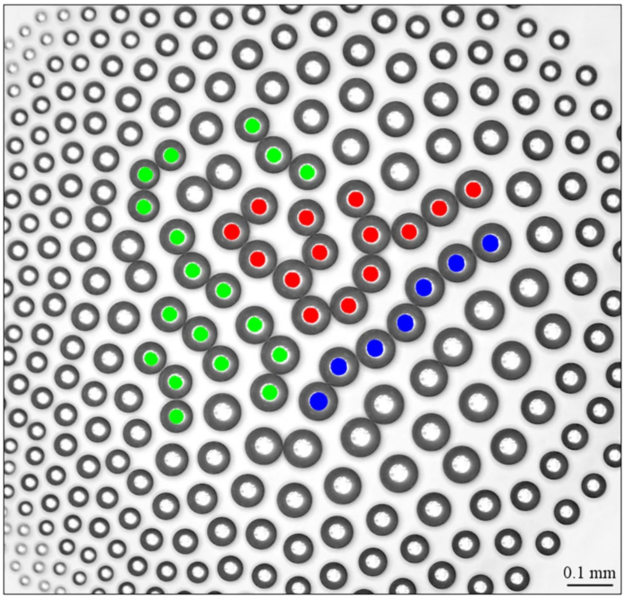


Fig. 7 A part of an asymmetric cluster containing growing branched chains of water droplets

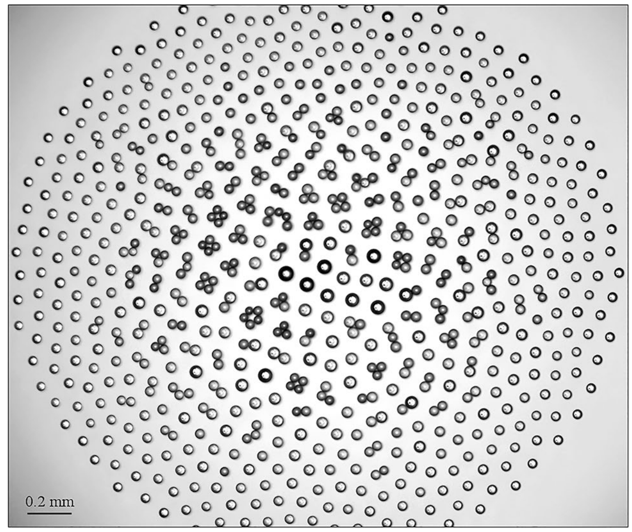


Fig. 8 A fragment of the axisymmetric hierarchical cluster of water droplets

the cluster and the only possible path for it is between the water droplets in the center of the cluster. In a small cluster, the droplets could simply move apart, letting in water vapor or its mixture with heated air to flow through the cluster. However, this is not possible in a large cluster. As a result, some of the large droplets close (but do not merge) with each other, forming branching chains. The average width of the gaps between the droplet chains in a chain cluster is greater than the distance between droplets in a hexagonal cluster. Therefore, the hydraulic drag of the central part of the chain cluster is less, which makes it possible for the increased amount of vapor-air mixture to flow.

Interestingly, the transition from a hexagonal to a chain cluster, like other second-order phase transitions, is reversible. For example, under external infrared irradiation, the chain cluster transforms it into the former hexagonal cluster [24]. As an additional illustration, Fig. 7 shows a fragment of a growing chain cluster with chains of droplets highlighted in different colors for clarity.

4.4 An advanced experimental procedure and hierarchical clusters

After several years of research, the recently discovered fundamentally new hierarchical cluster structure [26] came as a big surprise. In the central region of the hierarchical cluster, groups of several almost merging droplets are formed (Fig. 8), separated by a submicron layer of gas. Interestingly, both aerodynamic and electrostatic forces are responsible for droplet interaction within each group. Groups of droplets with different electric charges are constantly rearranged, droplets are exchanged, and individual droplets merge. At the same time, the outer layers of the hierarchical cluster retain a stable hexagonal structure. The droplet merging effect characteristic of the hierarchical cluster is of interest for microbiological and chemical experiments, since it is expected to allow in situ studies of merging the droplets of different chemical compositions.

5 Conclusions

The effect of the temperature profile of the water layer surface on the formation and structure of the droplet cluster was studied experimentally. It was shown that a local temperature maximum is a necessary condition for the formation of a self-assembled cluster. The Voronoi entropy, which characterizes the degree of orderliness of the hexagonal cluster structure, increases in proportion to the temperature difference between the center of the hot spot and the periphery of the water layer. Laboratory experiments showed that the distance between cluster droplets increases as the hot spot area on the water surface decreases. Analysis of conditions for formation of a cluster of levitating droplets is completed by a statement of a physical model of cluster evolution based on numerous laboratory observations. As before, this model is based on representations of gas dynamics. At the same time, it is noted that when analyzing the behavior of droplet aggregates in recently studied hierarchical clusters, it is necessary to take into account electrostatic forces.

The new images of various droplet clusters so different in shape and structure illustrated the diversity of the observed clusters. The experimental results presented in the paper are important for a better understanding the unusual behavior of diverse self-assembled droplet clusters. This may also be needed for further practical use of levitating droplets as promising microreactors in biochemical studies.

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Author contributions All authors contributed to the study conception. The first author designed the experimental setup and directed all experiments. DVS was engaged in the automation of measurements and participated in the experiments. AAF and LAD prepared the draft manuscript. All authors read and approved the final manuscript.

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Declarations

Competing interests The authors have no relevant financial and non-financial competing interests.

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